



Supporting Information

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**Isolated Fe Single Atomic Sites Anchored on Highly Steady
Hollow Graphene Nanospheres as an Efficient Electrocatalyst
for the Oxygen Reduction Reaction**

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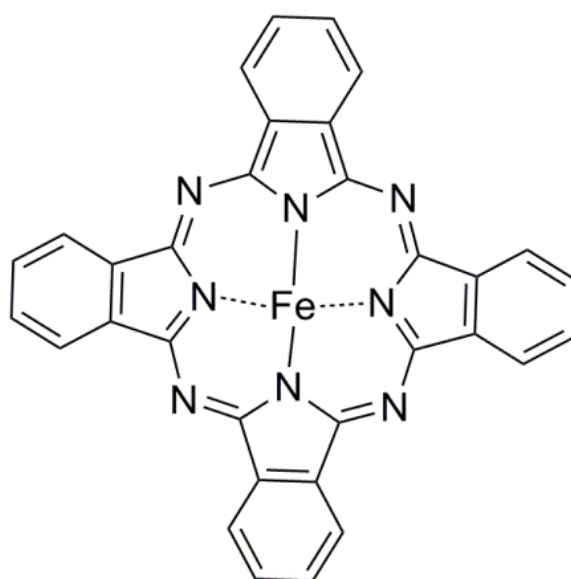


Figure S1 Molecular structure of iron phthalocyanine (FePc).

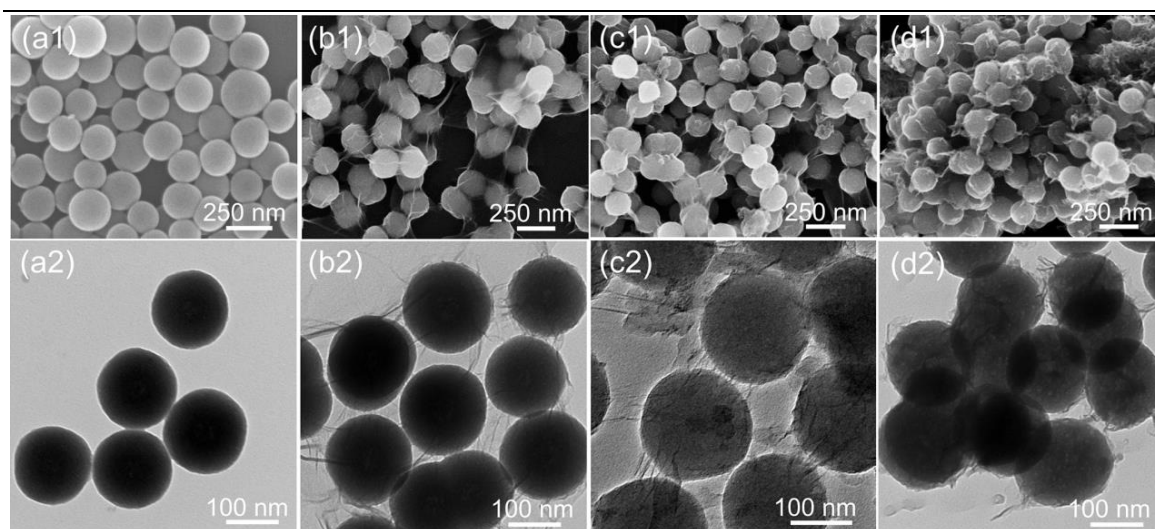


Figure S2 TEM and SEM images of the intermediate products. (a1)-(a2) SiO₂ nanospheres, (b1)-(b2) SiO₂@GO nanospheres, (c1)-(c2) SiO₂@GO/FePc nanospheres, and (d1)-(d2) SiO₂@rGO/Fe-N_x ISAs nanospheres.

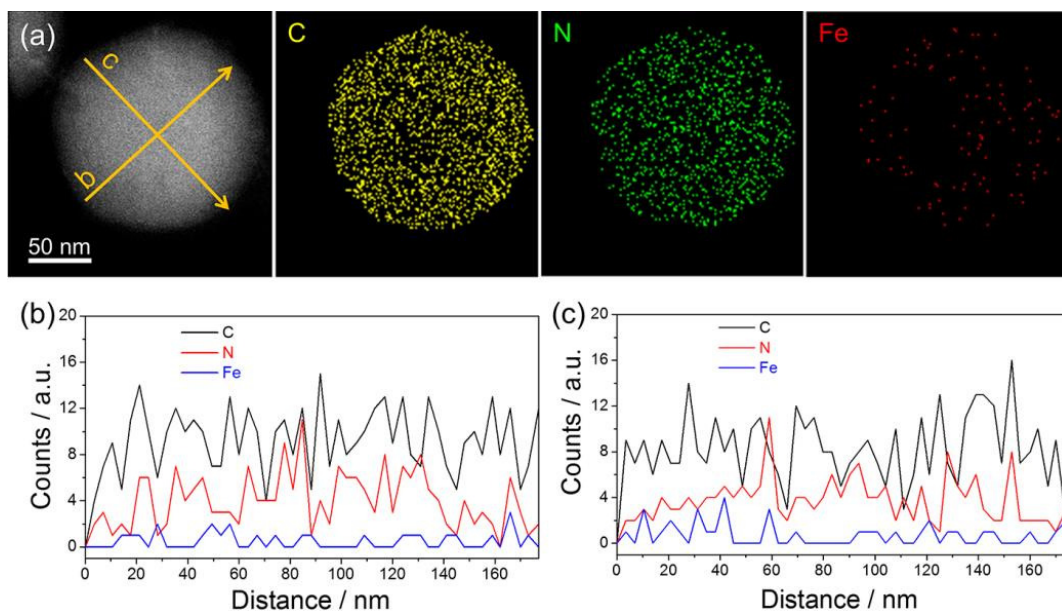


Figure S3 (a) HAADF-STEM image and elemental mapping images, and (b)-(c) EDX line-scan profiles of the SiO₂@GO/FePc nanospheres.

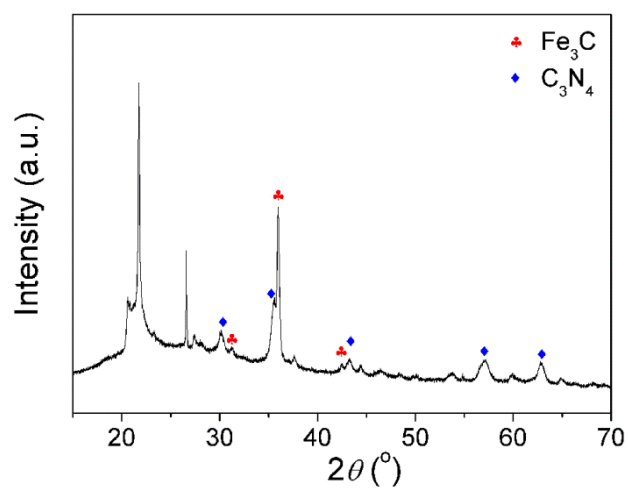


Figure S4 XRD pattern of the product derived from the direct pyrolysis of FePc molecules.

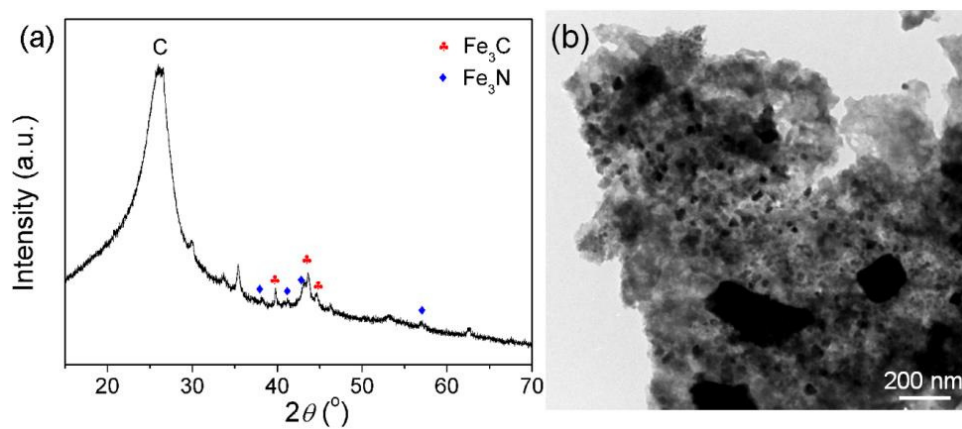


Figure S5 (a) XRD pattern and (b) TEM image of the FePc/GO nanosheets-derived product (denoted as Fe-N_x/GNSs).

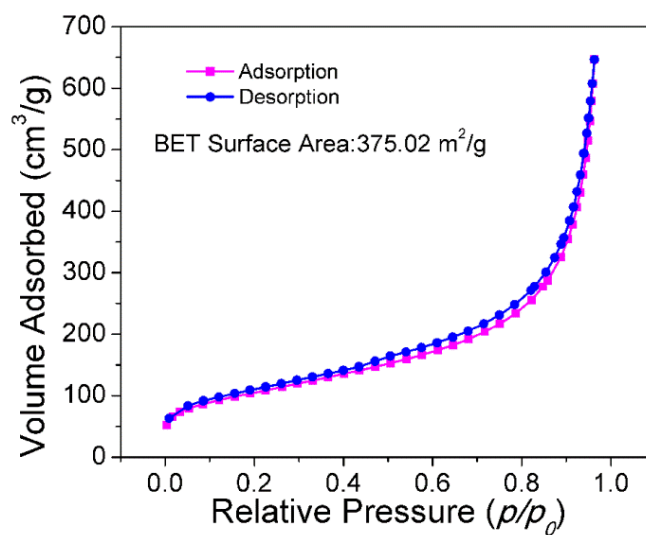


Figure S6 N₂ adsorption-desorption isotherms of Fe-N_x ISAs/GHSs.

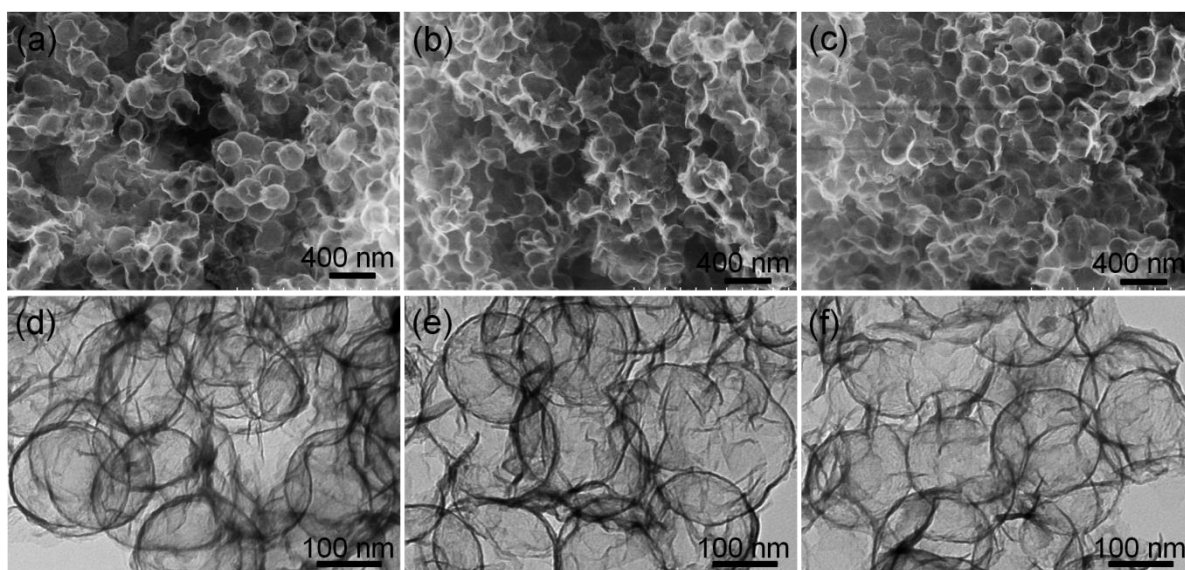


Figure S7 TEM and SEM images of the products prepared at different temperatures. (a)-(d) 600 °C, (b)-(e) 800 °C and (c)-(f) 900 °C.

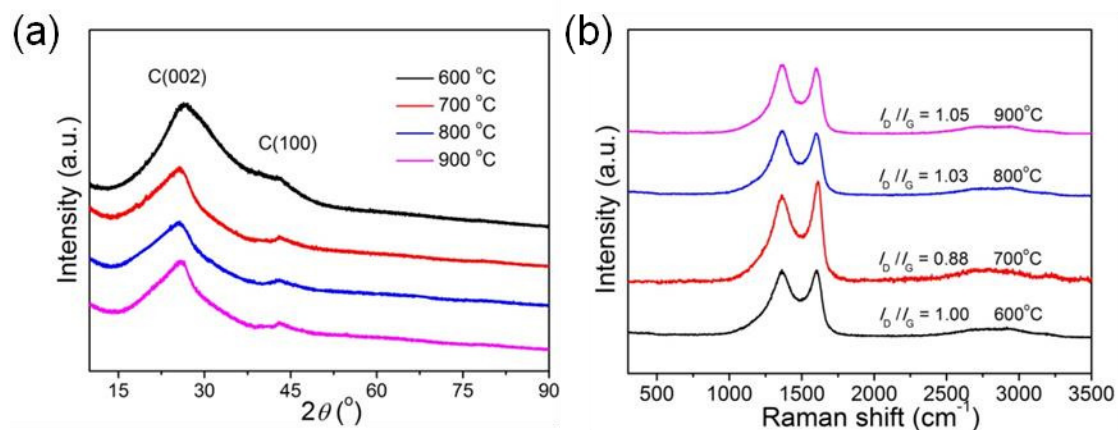


Figure S8 (a) XRD patterns and (b) Raman spectra of the products prepared at different pyrolysis temperature.

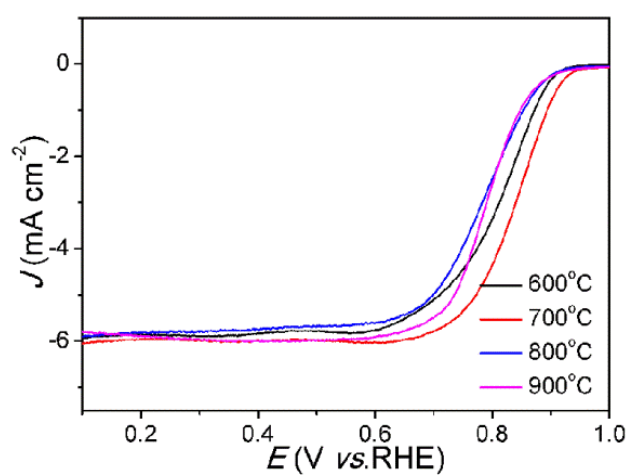


Figure S9 ORR polarization curves of the Fe-N_x ISAs/GHSs prepared at different pyrolysis temperatures in O₂-saturated 0.1 M KOH solution at a sweep rate of 5 mV s⁻¹ at 1600 rpm.

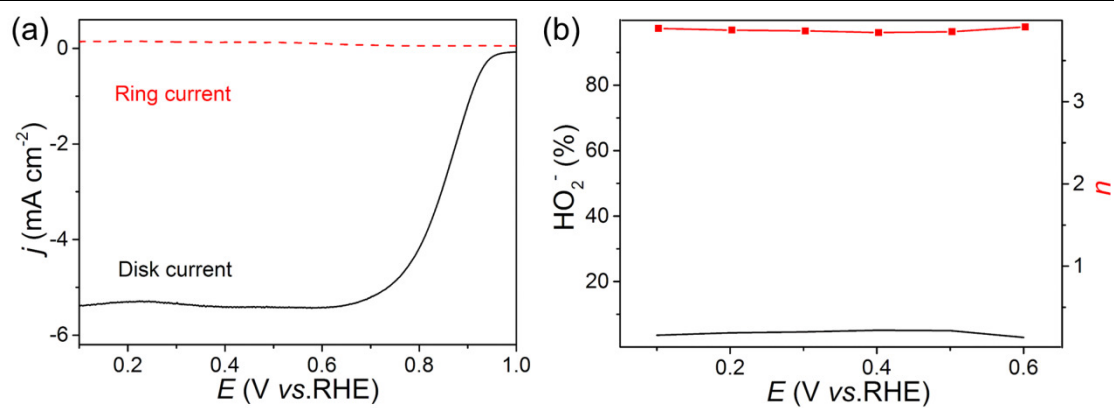


Figure S10 (a) RRDE test of the Fe-N_x ISAs/GHSs in O₂-saturated 0.1 M KOH solution at a sweep rate of 5 mV s⁻¹ at 1600 rpm. (b) HO₂⁻ yield and electrons transfer number *n* of the Fe-N_x ISAs/GHSs during the RRDE test.

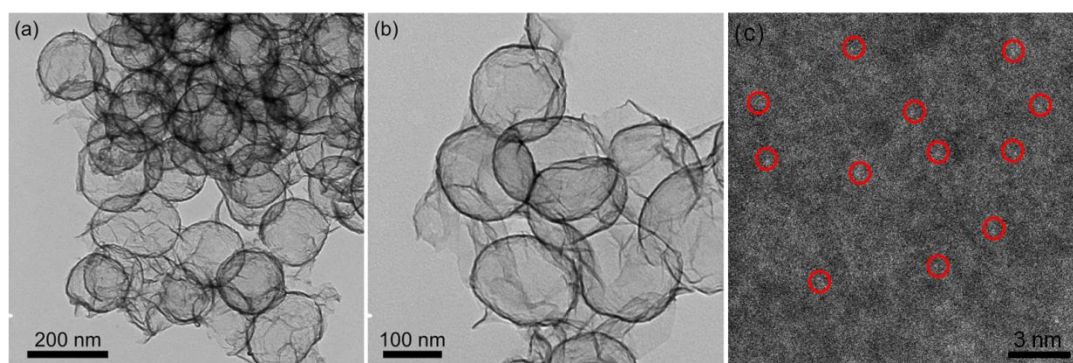


Figure S11 (a)-(b) TEM images and (c) aberration corrected HAADF-STEM image of the Fe-N_x ISAs/GHSs after the stability test.

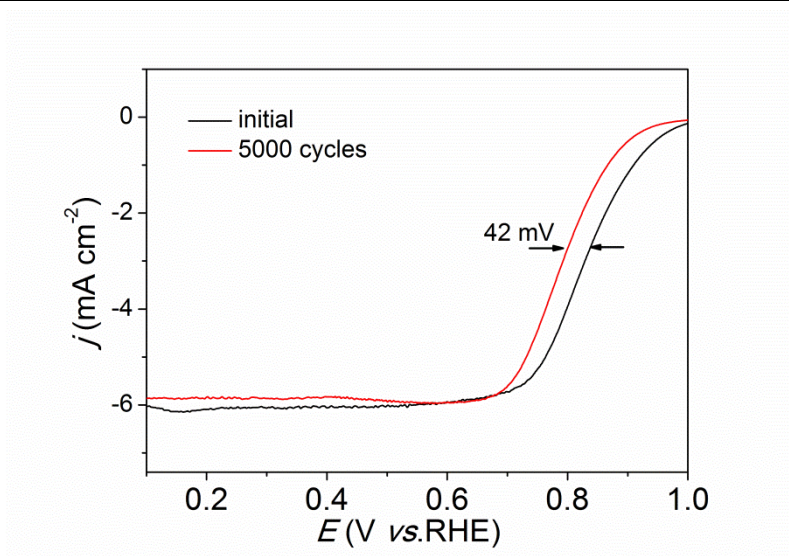


Figure S12 ORR polarization curves of the Pt/C before and after 5000 cycles.

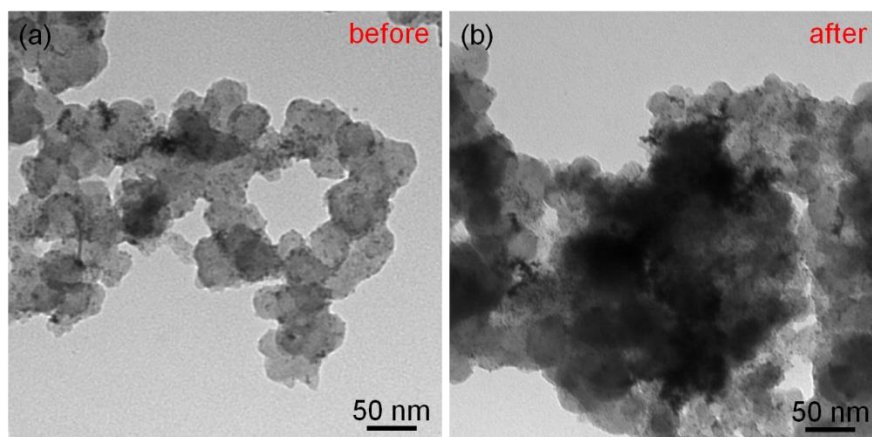


Figure S13 TEM images of the Pt/C (a) before and (b) after the stability test.

Table S1. Comparison of the ORR performance of the synthesized Fe-N_x ISAs/GHSs with some previously reported non-precious catalysts in KOH solution.

Number	Electrocatalysts	E_{onset} / V vs. RHE	$E_{1/2}$ / V vs. RHE	Ref
	Fe-N _x ISAs/GHSs	1.05	0.87	This work
1	p-Fe-N-CNFs	0.94	0.82	Energy Environ. Sci. 2018, DOI: 10.1039/c8ee00673c
2	FeSAs/PTF-600	1.01	0.87	ACS Energy Lett. 2018, 3, 883.
3	Atomic Fe-NGM/C-Fe	1.05	0.86	Chem. Mater. 2017, 29, 9915.
4	Fe-N-doped DSC	1.025	0.833	ACS Nano 2018, 12, 208.
5	pCNT@Fe@GL/CNF	N. A.	0.811	Adv. Mater. 2017, 1606534.
6	Fe-N-CNTAs-5-900	0.970	0.880	Small 2017, 13, 1603407.
7	Fe-N/MC@0.1	0.990	0.850	ChemCatChem 2015, 7, 2937.
8	Fe-ISAs/CN	0.990	0.900	Angew. Chem. Int. Ed. 2017, 129, 7041.
9	Graphene-like macroporous Fe-N-C Catalyst	N. A.	0.88	ACS Catal. 2017, 7, 6144.
10	Fe,N-doped carbon nanofibers	0.98	0.83	Chem. Mater. 2017, 29, 5617.
11	Fe/N-doped carbon nanofibers	0.88	0.79	Small 2016, 12, 6398.
12	Fe-N-doped mesoporous carbon microspheres	1.027	0.86	Adv. Mater. 2016, 28, 7948.
13	FeCo, N-codoped Porous Carbon Networks	1.050	0.88	Small 2016, 12, 4193.
14	Fe ₂ N/mesoporous N-doped graphitic carbon spheres	0.95	0.87	Nano Energy 2016, 24, 121.

15	Fe ₃ C nanoparticle embedded mesoporous carbon	1.02	0.86	Small 2016, 12, 5414.
16	C-FeZIF-900-0.84	0.950	0.860	ACS Appl. Mater. Interface. 2017, 9, 9699.
17	S,N-Fe/N/C-CNT	0.960	0.850	Angew. Chem. Int. Ed. 2017, 56, 610.
18	Fe-N/C-700 nanosheets	0.956	0.84	Small 2016, 12, 5710.